



Lawrence Livermore National Laboratory  
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FAX: (510) 423-6785

January 13, 1995

**TO: Elaine Gallin**

**TELE NO: (301) 903-2105**

**FAX NO: (301) 903-1413**

**FROM: Joseph H. Shinn**

**TELE NO: (510) 422-6806**

**SUBJECT: Status of LLNL Studies at Palomares, Spain.**

**MESSAGE:** Bill Robison asked me to pass along information about the status of our Studies at Palomares, Spain. I thought the best way was to include a March 2, 1994, progress report memo to Bill, as well as my 1993 Foreign Trip Report. As you know, all progress on this study stopped at the beginning of FY 1994. All the air samples and magnetic media records have been preserved, pending reinstatement of funding. If I can be of further assistance, please don't hesitate to call. I will be out of town next week, but back in my office on 23 January.

**We are transmitting 14 pages (including this cover sheet). If the transmittal is not complete, please phone (510) 422-3885.**

*Interdepartmental letterhead*

Mail Station L- 453

HEALTH &amp; ECOLOGICAL ASSESSMENT DIVISION

Ext: 2-6806

March 2, 1994

To: Bill Robison  
From: Joe Shinn  
Subject: Status of 1993 Palomares Study

The Palomares part of our Plutonium Pathway Analysis program remains inactive because of limitations in funding this fiscal year. For your consideration in finding a means to continue the Palomares project, I am providing the following report of progress and needs for that effort.

The purpose of the Palomares study is to assess the Pu source term of exposure to residents of Spain in the vicinity of the 1966 non-nuclear explosion of two nuclear weapons dropped during a mid-air collision. The study intends to supplement and assist the efforts of the Spanish national laboratory, Centro de Investigaciones Energeticas Medioambientales y Tecnologicas (CIEMAT), and is especially important in view of recent actions by the EPA to set standards for transuranic contaminants in soil. In 1993, we exchanged visits of analytical facilities with CIEMAT, exchanged Quality Assurance Standard Samples, and collaborated in a special resuspension study near the ground zero of one device to evaluate the inhalation pathway.

We successfully conducted a field reinvestigation (site 2-0 in Palomares) that was designed to collect the plutonium aerosol and meteorological data necessary for assessment of potential inhalation exposures and the plutonium aerosol emission rate. The site has been continuously monitored by CIEMAT.

To date, micrometeorological data have been analyzed to determine the diffusion coefficient so that the Pu-aerosol emission rate from the soil by resuspension could be determined, but the vertical-profile air samples (VA) have not been analyzed for Pu. There are 16 nighttime and 16 daytime VA samples waiting radiochemical analysis. Aerosol-Pu size distributions were sampled using cascade impactors. The Pu-activity on each impactor separation stage have not been determined. There are 20 impactor samples awaiting radiochemical analysis. High volume air samplers were operated to determine the total suspended particulates (mass loading), of the relatively dusty air. We found an average of  $79 \mu\text{g}/\text{m}^3$  mass loading of total suspended particulates, which is a high value and indicates that the air is very dusty. The specific activity of these samples (Bq/g), however, has not yet been determined. There are 8 high volume samples awaiting radiochemical analysis.

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Some air sample analyses have been completed, however. Ultra high volume samples (6), and isokinetic air samples (2) have gone through radiochemistry. We found that during the sampling period, the median Pu concentration in air determined from our ultra high volume air samplers was averaging about  $50 \mu\text{Bq}/\text{m}^3$ , or about 100 to 300 times world-wide background. Judging from these results, the enhancement factor at the site is much different from expected, and this could have considerable effect on the dose per unit of soil Pu, a factor needed in risk assessments and of importance to EPA in setting standards for Pu in soil. That is, the specific Pu activity (Bq/g) in the aerosols is two orders of magnitude lower than the Pu activity in the soil below. More information about this will be available when all sample results are known.

There are a total of 60 samples not yet analyzed radiochemically. In addition, we are recommending some follow-on work based on our experience there. Subsequent field trips should do further air sampling with high volume air samplers to determine the down-gradient values of the enhancement factor. The question to be answered is whether the value at site 2-0 is representative. If the enhancement factor increases with distance from the ground zero, it could be that exposures are greater per unit of soil activity as one approaches the heavily-farmed area. One would expect this if the Pu particle size in the soil decreases further out. In Nevada, explosive Pu releases had much higher enhancement factors.

We were generally pleased to find that CIEMAT does high-quality work. It would help them, however, if their Pu-exposure modelling capability was more practical, that is, converted from an academic, research orientation into an operational capability that could be used to estimate exposures for a set of real cases at Palomares. We propose to advise and assist them to do that.

Please advise me about our options with the Palomares project. We would like to complete this phase of the Pu Pathway Analysis program in a timely way.

**FOREIGN TRIP REPORT****JOSEPH H. SHINN, CLEO O. FRY, JAMES STEPHEN JOHNSON****HEALTH AND ECOLOGICAL ASSESSMENT DIVISION  
LAWRENCE LIVERMORE NATIONAL LABORATORY  
UNIVERSITY OF CALIFORNIA  
LIVERMORE, CA 94550****CONTRACT NO. W-7405-ENG-48****JULY 19, 1993****SECTION A. SUMMARY****a. Travelers: Joseph H. Shinn, Cleo O. Fry, and James Stephen Johnson**

Trip leader: Joseph Shinn, PhD      Telephone: 510/422-6806  
Health and Ecological Assessment Division, LLNL

**b. Dates and Destinations:**

June 12, 1993 - July 4, 1993. Madrid and Palomares, Spain

**c. Purpose of the Trip**

The purpose of the trip was to carry out a field research campaign, or *Resuspension Study*, sponsored by DOE-EH, involving investigators from a Spanish national laboratory, and under a plan previously worked out and approved. The trip involved the installation and operation of specialized air monitoring equipment belonging to DOE, and shipped from LLNL.

**d. Summary of the Trip**

The travelers, Joseph Shinn, Cleo Fry, and J. Stephen Johnson, are a team of environmental scientists who conducted an investigation in Palomares, Spain, of the potential inhalation exposure to plutonium (Pu) aerosols. The Pu particles in the soil originated from a B-52 aircraft accident in 1966, when two nuclear weapons exploded and burned in the farm community of Palomares. This study was a cooperative effort with the *Centro De Investigaciones Energeticas, Medioambientales Y Tecnologicas* (CIEMAT) of Madrid. As defined in the joint *Resuspension Study Plan*, the technical purpose was to verify the resuspension factor and determine the resuspension rate during a two-week period in June. The travelers installed and operated the equipment, and decontaminated and packed the equipment for return to LLNL, during the period 12 June to 4 July, 1993.

**SECTION B: TRIP REPORT****a. Purpose of the Trip:**

The purpose of the trip was to carry out research with CIEMAT, as previously agreed in April, 1993. (See attached copy of *Visit Report for C. E. Iranzo, and A. Espinosa, CIEMAT, April 26-30, 1993, HEA Division, LLNL.*) Two parts of the Cooperative Program for CIEMAT and LLNL, (memo of Bill Robison included in the above visit report) were carried out. First, the initial part of the Quality Control Procedures, involved a visit to CIEMAT, Madrid, and inspection of their gamma- and alpha-analysis equipment and procedures. This visit was also reported in the Foreign Trip Report of William L. Robison, July 1, 1993. Second, Part C of the Cooperative Program, Resuspension Studies, was carried out in Palomares, Spain, as specified in the Resuspension Study Plan (copy included in the above visit report attached).

**b. Summary of Activities:**

The travelers arrived in Madrid on 13 June, 1993. Messrs. Johnson and Fry traveled overland to Palomares, Spain, on 14 June, 1993, in a vehicle provided by CIEMAT. Dr. Shinn visited CIEMAT 14 and 15 June, 1993, with Dr. William Robison, and went through the laboratories to see their analytical capabilities, evaluate their sample processing procedures, and to arrange for the exchange of soil and vegetation samples for quality control purposes. In addition, Dr. Shinn was briefed on the meteorological modelling and monitoring data processing which was part of the Palomares project. On 15 June, 1993, Drs. Shinn and Robison traveled by air to Palomares in the southeastern corner of Spain. Beginning 16 June 1993, the resuspension study equipment was installed, until 18 June 1993, when Mr. Johnson and Dr. Robison departed as planned to return to USA. Dr. Shinn and Mr. Fry continued to operate the equipment as planned until 1 July 1993, when the equipment was decontaminated, packaged, and placed on a CIEMAT truck for return to Madrid, on 2 July, 1993. Dr. Shinn and Mr. Fry traveled to Madrid, where they departed for USA on 4 July, 1993. The Resuspension Study Plan was followed exactly. The only significant change in operation was the necessity of renting a 50 KVA generator by CIEMAT, because the power line into the field site proved inadequate to carry the electrical load, even though LLNL advised CIEMAT of its electrical requirements prior to travel.

**c. Travelers' Roles**

Dr. Shinn was the trip leader with responsibilities to carry out the Resuspension Study, and to prepare for Quality Control Procedures that are relevant to the potential inhalation exposure in the human health assessment. Cleo Fry was the coordinator for all instrumentation, including calibration and preparation, packing, arranging transportation, installation, electrical power, and operation. He installed and operated an automatic micrometeorological

station, a boundary layer parameter station, 4 cascade impactor units, 4 high-volume air samplers, 2 vertical array air sampler units, 1 ultra high-volume air sampler, 1 AQ-10 air mass-loading monitor, and 1 isokinetic particle counter unit. James Stephen Johnson was a specialist in setup and operation of cascade impactor and high-volume air sampling units, who assisted in the installation of equipment and then returned to LLNL. Dr. Shinn and Cleo Fry carried out the experimental phase of the Resuspension Study, beginning 19 June and continuing to 30 June.

CIEMAT provided drivers and assistance, and an interpreter, so that we were accompanied at all times by CIEMAT personnel.

#### d. Recommendations

Our relationship with the Spanish national laboratory, CIEMAT, has developed into a professional collegiality. We have assisted and advised their personnel with regard to long-term monitoring of the Pu contamination, and the associated human health assessments. Their personnel have done excellent work in characterizing the extent of contamination, characterizing the ingestion pathway, monitoring suspended Pu, and monitoring the local population. Their analytical capabilities in Madrid appear to be excellent. Our joint study of the inhalation exposure pathway and joint quality assurance will lead to collaborative publications and possibly a long-term commitment of effort. We recommend that further collaborative efforts be continued, for both professional reasons and because of the scientific importance of the Palomares data. There is no better opportunity anywhere in the world, to study a population where a highly complex society with intensive agricultural operations, exists in proximity to Pu contamination without governmental control.

#### e. Information Pertinent to US Energy Postures.

There is only indirect relationship of these environmental studies to the energy development in the US, in its tangential relation to nuclear power plant safety.

#### f. Security-Related Concerns.

None. We had no need to discuss classified information and there was apparently no interest shown by Spanish nationals in any part of our work except the environmental studies in our collaborative effort.

### **SECTION C: APPENDIX**

#### a. Itinerary

Travelers Leave San Francisco 0825 6/12/93

Travelers Arrive Madrid 0730 6/13/93

Johnson and Fry depart Madrid 1000 6/14/93

Johnson and Fry arrive Palomares area, 1900 6/14/93  
Shinn, business in Madrid 6/14-15/93  
Shinn, depart Madrid 1950 6/15/93  
Shinn, arrive Palomares area, 2230 6/15/93  
Johnson, business in Palomares area 6/15-18/93  
Shinn and Fry, business in Palomares area 6/15-7/2/93  
Johnson depart Palomares area 2130 6/18/93  
Johnson arrive Madrid 2230 6/18/93  
Johnson depart Madrid 1235 6/19/93  
Johnson arrive San Francisco 2130 6/19/93  
Shinn and Fry depart Palomares area 2130 7/2/93  
Shinn and Fry arrive Madrid 2230 7/2/93  
Shinn and Fry depart Madrid 1235 7/4/93  
Shinn and Fry arrive San Francisco 2130 7/4/93

b. List of Persons Contacted:

Dr. Emma Iranzo, CIEMAT Institute of Radiation Protection and Environment  
Dr. Ascuncion Espinoza, CIEMAT Institute of RPE  
Dr. Jose Gutierrez, Director, CIEMAT Institute of RPE  
Dr. Francisco Mingot, Director, CIEMAT Environmental  
Dr. Emilio Iranzo, Former Director, CIEMAT Institute of RPE  
Dr. Javier Martinez, CIEMAT Institute of RPE

c. Literature Acquired:

None

d. Attachments

LLNL Visit Report for C. E. Iranzo and A. Espinosa, CIEMAT, April 26-30, 1993.  
Memo, Cooperative Program for CIEMAT and LLNL, April 7, 1993.  
Resuspension Study Plan

VISIT REPORT FOR C. E. IRANZO AND A. ESPINOSA,  
CIEMAT (SPAIN), APRIL 26-30, 1993, HEA DIVISION, LLNL.

The visitors were Concepcion-Emma Iranzo, and Asuncion Espinosa of the *Centro De Investigaciones Energeticas, Medioambientales Y Tecnologicas* (CIEMAT), Madrid, Spain. These scientists are health physicists who have been responsible for investigations (Project Indalo) of the results of a nuclear accident in 1966, when a U.S. B-52 aircraft exploded in mid-air and radioactive plutonium contaminated approximately one square mile near Palomares, Spain. The purpose of the visit was to plan a joint field investigation of plutonium aerosol resuspension at the site in June, 1993, funded by DOE-EH. In addition, analytic methodologies, experimental protocols, and exchanges of standard samples for quality control were agreed upon. The visit to LLNL took place April 26-30, 1992.

The host of the visit was Dr. Joseph H. Shinn. Members of the LLNL staff who participated in discussions were Dr. Victor Noshin, Kai Wong, Jim Brunk, Cleo Fry, Jim Stephens Johnson, Frank Gouveia, Carol Stoker, Cynthia Conrado, and Dr. Lynn Anspaugh.

Discussions were held on the explanation of organizations and programs of CIEMAT and LLNL, analytic methods and protocols of gamma and alpha spectroscopy of environmental samples, review of data base management, sample chain-of-custody, and quality control procedures. Plans were made and a Resuspension Study Plan for the next field campaign in Palomares, Spain, 14 June to 3 July 1993. A budget for both CIEMAT and LLNL participation in the field campaign was drawn up. Future collaborative work was outlined and discussed without definite commitments.

Tours were provided to the Spanish visitors of the environmental gamma spectroscopy facility (B-379), alpha spectroscopy laboratory (B-378), air sampling and meteorology laboratory (B-412), soil processing laboratory (B-591), and sample freeze-drying laboratory (B-412). Discussions and meetings were held in the conference room of Health and Ecological Assessment Division (B-3703).

Agreements were made to carry out the quality control procedures and resuspension studies recorded in a memorandum of April 7, 1993, from Bill Robison, HEA Division Leader, to C. E. Iranzo, CIEMAT visitor, see copy attached. We agreed also to pursue future collaborative studies outlined in the above-mentioned memorandum. We jointly prepared a

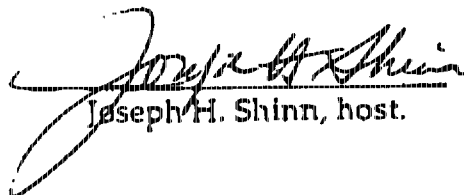


draft Resuspension Study Plan as a basis for our next field campaign, see copy attached. We have a few questions to be answered regarding available electrical power and travel arrangements, which will be satisfactorily completed by exchange of FAXes. We agreed on a budget total of \$74K for CIEMAT and \$120K for LLNL to be requested of DOE for the joint field campaign.

Written material provided to the visitors were, (1) copies of the referenced (attached) memorandum of April 7, 1993, (2) copies of the Resuspension Study Plan (attached) and a written summary of the meetings, (3) a copy of a technical (unclassified) report by Joseph Shinn and Frank Gouveia, *The "Footprint" Area Influencing a High-Volume Air Sampler* UCRL-ID-112181, October 1992, and (4) two graphs analyzing the cumulative frequency distribution of CIEMAT air monitoring data of plutonium aerosols from the Palomares 2-1 and P sites for the purpose of planning the field campaign.

The benefits to LLNL and DOE of this visit will be realized in a well-prepared and successful field campaign to be accomplished in June, 1993, with DOE sponsorship. Results of the visit will be briefed to DOE-EH by Dr. Bill Robison.

To the best of my knowledge, no classified or unauthorized sensitive subject has been disclosed to the foreign national visitors.

  
Joseph H. Shinn, host.

May 7, 1993.



**Lawrence Livermore National Laboratory**  
**ENVIRONMENTAL SCIENCES DIVISION**

April 07, 1993

To: Emma Iranzo  
From: Bill Robison  
Subject: Cooperative Program for CIEMAT and LLNL

**Draft Protocols for Joint Work between CIEMAT and LLNL**

Each agency will provide split samples to be exchanged by October 15, 1993. Analysis will be completed by February 15, 1994. Final reports will be completed by April 20, 1994.

**A. Quality Control Procedures: Exchange of Soil Samples for Pu and Am Analysis.**

Ten (10) soil samples will initially be exchanged between CIEMAT and LLNL. CIEMAT will supply 10 soil samples of their choosing from Palomares; LLNL will supply 10 coral soil samples of their choosing from the Marshall Islands.

The coral soil samples from LLNL will be dried at 100°C to constant weight, ballmilled to a fine-powder consistency, ashed at 450°C, and a 100 g aliquant sent to CIEMAT. An estimate of the range of the Pu and Cs concentration will be provided for each sample to help separate hi-concentration samples from low-concentration samples and to help the analysts select an appropriate sample size for analysis.

The Palomares soil samples from CIEMAT will be dried to constant weight, ground in a mortar and pestle, and a 100 g aliquant sent to LLNL along with an estimate of Pu concentration.

We have our current soil permits changed to cover samples from Spain as well as the Marshall Islands. We will forward these permits to you; they are to be placed on the shipping container you will send us and the samples will then be allowed through the U.S. Department of Agriculture (USDA) Inspection and Customs.

## Draft Profotols for CIEMAT and LLNL

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**B. Quality Control Procedures: Exchange of Vegetation Samples for Pu and Am Analysis**

Ten (10) homogenized vegetation samples will initially be exchanged between CIEMAT and LLNL. CIEMAT will supply 10 vegetation samples of their choosing as will LLNL. Each vegetation sample should be between 1 and 2 kg dry weight. We will exchange at least three replicate vegetation samples with associated soil profiles in the root zone so that we can evaluate the concentration ratios (CR's) for food crops (these soil samples are part of the 10 soil samples mentioned in Part A). Fresh dry and ash weight ratios will be provided. The vegetation samples will be supplied in the form of ashed samples.

We have our vegetation permits changed to include samples from Spain. As with the soils, this will allow passage through the USDA and Customs inspections. We will forward the permits to you.

**C. Resuspension Studies**

LLNL is planning to support CIEMAT in additional resuspension work at Palomares. We will supply additional equipment—hi-volume air samplers, cascade impactors, ultra-hi volume air samplers, meteorological towers, etc., and personnel. We will plan with CIEMAT during your visit to LLNL in April the details of what equipment and how many people from LLNL will participate in the Palomares study. Air filters from this research effort will be split for analysis by both CIEMAT and LLNL. A minimum of twenty-five percent of the filters will be divided with one half going to each group.

**D. Future Contact**

CIEMAT personnel will visit LLNL in late April 1993. At that time there will be detailed planning of the resuspension work to be done at Palomares in the summer of 1993. LLNL personnel will then visit the CIEMAT laboratory in Madrid and travel to Palomares with CIEMAT staff in June 1993.

Additional soil and vegetation samples for intercomparison will be exchanged between CIEMAT and LLNL in 1994. LLNL will also send a bulk coral soil sample to CIEMAT in 1994 for particle size analysis.

Future field work at Palomares and possible joint field trip to the Marshall Islands will be discussed in our meeting in Madrid in June 1993. These co-operative projects will be conducted over the next few years at a pace that is consistent with the on-going commitments of both CIEMAT and LLNL.

Draft Profotols for CIEMAT and LLNL

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E. Budget Requirements

CIEMAT should forward any budget requests for this cooperative effort directly to Dr. Harry Pettengill in Environment, Safety and Health (ES&H) in the Department of Energy (DOE).

# DRAFT

## Resuspension Study Plan

LLNL and CIEMAT will carry out a special resuspension study at Palomares, Zone 2-0, north of impact point 2. The purpose of this study will be to verify the resuspension factor and determine the resuspension rate through joint efforts during a two-week period in June 1993. We will determine the representativeness of this time period by comparison to existing, local meteorological and air monitoring data.

CIEMAT will provide:

- (1) Meteorological data from a nearby tower (wind speed and direction), and air monitoring data ( $^{239+240}\text{Pu}$ , and  $^{241}\text{Am}$ ) from Station 2-1;
- (2) necessary soils data from historical sampling and FIDLER surveys of the resuspension study plot;
- (3) air sampling data from high-volume cascade-impactor air samplers at two heights (1.8 m and 3.0 m) in the plot during this study period;
- (4) electrical power sufficient for operation of all air samplers;
- (5) transportation and installation assistance to the LLNL team;
- (6) anticontamination support and decontamination services as needed.

LLNL will provide:

- (1) Micrometeorological data from a portable, solar-powered automatic weather station measuring wind speed/temperature profiles, and energy budget components (sensible heat flux, ground heat flux, and net thermal/solar radiant energy flux) up to a height of 2 m;
- (2) air sampling vertical profiles at four heights up to 2 m with 8 units sampling at about  $20 \text{ m}^3/\text{h}$ ;
- (3) air sampling vertical profiles at two heights (0.9 m and 1.8 m) with 4 high-volume units sampling at about  $67 \text{ m}^3/\text{h}$ ;
- (4) particle-size air sampling with 4 high-volume cascade impactors (Anderson, jet-type) at 1 m height operating at  $33 \text{ m}^3/\text{h}$ ;
- (5) particle-size analysis of mass-distribution in air with an optical particle counter utilizing an isokinetic wind-vane inlet;
- (6) air sampling for 24 h (or shorter) intervals using an ultra high-volume unit sampling at  $600 \text{ m}^3/\text{h}$ .

A minimum number of 12 air samples from LLNL units will be provided to CIEMAT (2 high-volume, 4 vertical array, 5 cascade impactors, 1 ultra high-volume). A minimum number of 12 air samples and maximum of 36 air samples will be analyzed by LLNL. The micro-meteorological results of estimation of particle diffusion coefficients corrected for atmospheric stability will be provided by LLNL using hourly-averaged data.

# DRAFT

**DRAFT****Resuspension Study Plan****2**

LLNL and CIEMAT will each analyze their (partitioned) air samples, and exchange results of analyses ( $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$ ) according to their established protocols. A joint publication will be prepared of estimates of resuspension factors, resuspension rates and particle-size analysis. Preliminary analysis results will be exchanged by October 15, 1993, and a preliminary draft report will be completed by January 30, 1994. Final reports will be completed by March 30, 1994.

**DRAFT**